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MAXIMUM SUPERCOOLING IN LIQUID ^{3}He-^{4}He MIXTURES NEAR THE TRICRITICAL POINT*

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Measurements of supercooling in liquid ^{3}He-^{4}He mixtures near the tricritical point are presented. The reduced temperature range $0.001 < \epsilon \equiv (1 - T/T_t) < 0.01$ was investigated for three different rates of cooling using a pressure-quench technique. For $\epsilon < 0.012$, the maximum supercooling was found to be a function of the cooling rate. Comparisons with data in organic binary mixtures are given.

1. INTRODUCTION

We have made quantitative measurements of the maximum supercooling δT_m in the superfluid phase of liquid ^{3}He-^{4}He mixtures at three different quench rates in the vicinity of the tricritical point T_t. A pressure quench technique (1) was used to produce supercooling in the mixture and the sudden attenuation in the intensity of a transmitted laser beam through the mixture was used to determine the cloud-point, which marks the limit of supercooling δT_m. Employing a similar technique, Alpern et al. (2) recently found that as the tricritical point is approached, i.e., as $\epsilon \equiv (1-T/T_t) \to 0$, the normalized maximum supercooling $\delta T_m/\Delta T$ tends to diverge. We use the definitions $\delta T_m \equiv T_{ps} - T_{cp}$ and $\Delta T \equiv T_t - T_{ps}$, where T_{ps} is the equilibrium phase separation temperature and T_{cp} is the measured cloud-point temperature. Such behavior of super-cooling near the critical point has been observed for organic binary mixtures (3), but Alpern et al. showed the divergence of $\delta T_m/\Delta T$ to be much stronger in the case of ^{3}He-^{4}He. They also pointed out that this feature of stronger divergence of the supercooling near T_t could be a peculiarity of tricritical systems. In this paper, we show that the degree of divergence of $\delta T_m/\Delta T$ as $\epsilon \to 0$ is a function of the quench rate, i.e., the rate of cooling; and if one compares the ^{3}He-^{4}He data with those of organic mixtures for equivalent quench rates, then no meaningful discrepancy is observed. We also show other similarities in the results between ^{3}He-^{4}He and other studies in organic binary mixtures (4).

The divergence of supercooling as $\epsilon \to 0$ can be explained in terms of critical slowing-down of diffusion processes (5,6). The experimentally determined cloud-point indicates the stage of phase-separation when the evolving minority-phase clusters or droplets have grown large enough to begin to scatter light strongly. Consequently, the cloud-point phenomena always lag in time behind the actual onset of homogeneous nucleation. Because of critical slowing down, this time-lag gets progressively larger as one approaches the critical point. Coupled with the finite quench rates used in actual experiments, this appears to indicate additional supercooling.

2. EXPERIMENTAL

The supercooling of the liquid ^{3}He-^{4}He mixture in the metastable region of the miscibility gap below the tricritical point was produced using a pressure quench technique (1). Pressurization of the ^{3}He-^{4}He mixture inside the sample cell was accomplished by pure ^{4}He actuating two horizontal bellows moving toward each other. A germanium resistance thermometer monitored the temperature T and an in-situ capacitive transducer with a resolution of 10^{-5} atm monitored the pressure P of the mixture. The intensity I_0 of the transmitted He-Ne laser beam, attenuated to approximately 0.1 mW before entering the cell, was measured using a sensitive photodiode. Different pressure quench rates were made possible by using several parallel and independently selectable fixed impedances in the pressure release line of the pressurization bellows outside the cryostat. Fig. 1 shows a typical pressure quench history. The cloud-point is indicated by the point where I_0 suddenly starts to decrease. The actual supercooling δT_m was evaluated from the pressure at the cloud-point and the well-known pressure dependence of the tricritical point (1,2).

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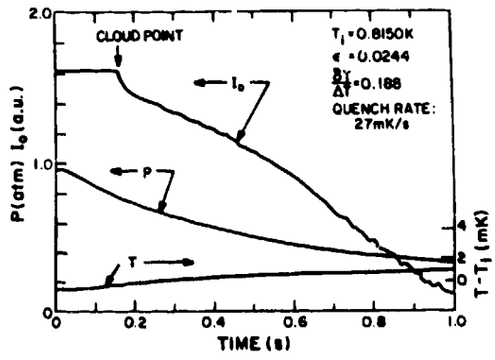

Figure 1. Time dependence of the pressure, temperature, and forward transmission intensity during a pressure-induced quench into the miscibility gap of a ^{3}He-^{4}He liquid mixture.

3. RESULTS AND DISCUSSION

We now discuss the results of our experiment to determine the homogeneous nucleation limit to supercooling. Three different rates of supercooling were used: 67, 27, and 12 mK/s. These are the average values of the cooling rates, determined from the pressure quench data, at the moment the light intensity I_o begins to drop (see Fig. 1). All the measurements were made in the superfluid phase of the mixture. Fig. 2 shows the reduced supercooling $\delta T_m/\Delta T$ as a function of ε on a log-log plot for the three different quench rates. The long-dashed line is the data of Ref. 4 for a quench rate of 30 mK/s in a lutidene-water mixture. The short-dashed line represents the data of Ref. 3 for lutidene-water using effective quench rates of less than 1 mK/s. The dotted line represents data in the ^{4}He phase of ^{3}He-^{4}He mixtures from Ref. 2 for quench rates greater than 150 mK/s. For values of $\varepsilon < 0.012$, the dependence on quench rate is obvious. All data can be fairly well represented by straight lines. At $\varepsilon > 0.012$, the quench rate dependence of the supercooling gradually diminishes and the $\delta T_m/\Delta T$ data become consistent with the prediction from classical nucleation theory (dot-dashed line). Similar behavior was observed by Strey et al. (4) in lutidene-water for three different quench rates used in their experiments (see Fig. 8 of Ref. 4). Considering the differing experimental techniques used (pressure quench vs. temperature quench) and the disparate thermodynamic properties of the two systems, no meaningful inconsistency can be inferred from data in organic mixtures in comparison to either our data or those of Benda et al. in ^{3}He-^{4}He, i.e., there is no unreasonably stronger divergence in the ^{3}He-^{4}He supercooling data as $\varepsilon \to 0$. However, the effects of the particular experimental techniques used,

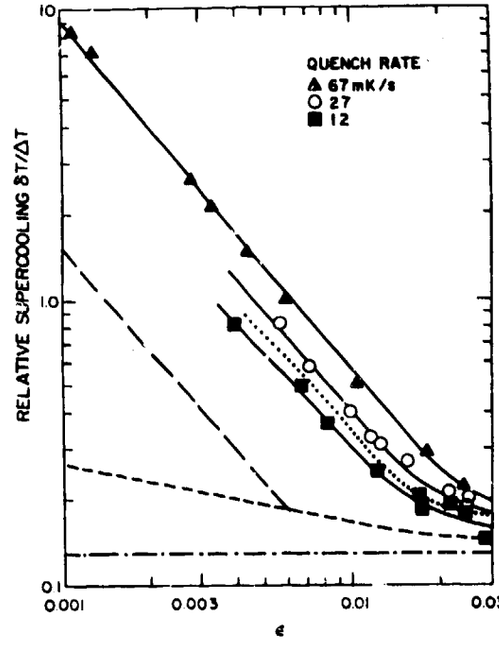

Figure 2. Relative supercooling vs reduced temperature for three different cooling rates in the superfluid phase of a ^{3}He-^{4}He mixture of tricritical composition.

such as pressure quench vs temperature quench, in determining the cloud-point, have not been adequately studied. Therefore, we do not feel that there is sufficient evidence to suggest that the divergence of supercooling near the tricritical point in ^{3}He-^{4}He is substantially different from that in organic mixtures near their critical points. In conclusion, we have shown that in ^{3}He-^{4}He mixtures, the anomolous maximum super-cooling is largely an artifact of the finite rates of cooling inherent to the experimental methods that have been utilized.

REFERENCES

(1) D. N. Sinha and J. K. Hoffer, Phys. Rev. Lett. 50 (1983) 515.
(2) P. Alpern, Th. Benda, and P. Leiderer, Phys. Rev. Lett. 49 (1982) 1267.
(3) A. J. Schwartz, S. Krishnamurthy, and W. I. Goldburg, Phys. Rev. 21A (1980) 1331.
(4) R. Strey, J. Wagner, and J. Woerman, Ber. Bunsenges. Phys. Chem. 86 (1982) 306.
(5) K. Binder and D. Stauffer, Adv. Phys. 25 (1976) 343.
(6) J. S. Langer and A. J. Schwartz, Phys. Rev. 21A (1980) 948.